# STRUCTURAL AND MICROWAVE DIELECTRIC PROPERTIES OF $Sr_{(1-X/2)}Na_XNb_2O_6$ FERROELECTRIC CERAMICS.

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**Abstract**

Strontium sodium niobate ceramics are prepared by solid state reaction method. The crystal structures of these ceramics are studied by X-ray powder diffraction method. These materials are found to crystallize in the tetragonal tungsten bronze type structure and the unit cell parameters are calculated. The microstructural features were studied using scanning electron microscope. The real and imaginary part of complex Permittivity of the sample is determined by cavity perturbation technique. It is observed that the real and imaginary part of complex Permittivity varies with frequency. The temperature coefficient of dielectric constant and resonant frequency is found to be very small.

## Introduction

Progress in science and technology relies heavily on the development of new materials. Among these, "Smart" or "intelligent" materials have an important role to play in the development of new sophisticated devices. Smart materials and structures incorporate one or more of the following features: (a) Sensors or actuators which are either embedded within a structural material or bonded to the surface of that material. (b) Control capabilities which permit the behaviour of the material to respond to an external stimulus according to prescribed functional relationship or control algorithm. At a more sophisticated level, such smart materials become intelligent when they have the ability to respond intelligently and autonomously to dynamically changing environmental conditions. Potential applications of these are wide spread and have excited interest in industrial, military, commercial, medical, automotive and aerospace fields.

Resonators made of dielectric materials are used in a number of applications in frequency bands covering audio, RF, microwave and optical frequencies. They are basic elements in filters, oscillators; antennas etc. Microwave dielectric resonators are widely used in mobile telephones, satellite broadcasting systems, wave guides, capacitors, and frequency filters. Oxide ceramics are crucial elements in these microwave devices and a thorough understanding of the crystal structure and physical properties is essential for future development.

Dielectric materials with high dielectric constant $\varepsilon_r$, high Q, and small temperature coefficient of resonant frequency $\tau_f$ are particularly required for developing microwave dielectric resonator[1]. The experimental characterization of dielectric materials suitable for resonant cavities has been studied. [2, 3]. To analyse the temperature stability one must know the properties of all the materials used in the construction of the cavity device. Dielectric resonators exhibiting high Q factor and very low temperature coefficient of resonant frequency have been [4, 5], they promise to shrink the size and cost of waveguide cavities. Developing a zero $\tau_f$ material is probably the most difficult aspect of research in microwave dielectrics. The challenge now a days is to find ways to tune $\tau_f$ without reducing $\varepsilon_r$ and maintaining quality factor (Q) [6]. When a particularly high Q is required, tantalates are used. The two main compounds in the market are $Ba(Mg_{1/3}Ta_{2/3})O_3$ (BMT) and $Ba(Zn_{1/3}Ta_{2/3})O_3$ (BZT). Although tantalates satisfy the high Q end of the market in terms of performance, $Ta_2O_3$ is expensive. Ideally, cheaper materials with equivalent $Q/\varepsilon$, values based on either niobates or titanates must be found. Structural and dielectric properties of $Sr_{1-x}Ba_xNb_2O_6$ [7], $Sr_{1-2x}K_xNb_2O_6$ [8] and $Ba_4Na_2Nb_2O_6$ [9] ceramic materials had been investigated by various researchers. But, physical and microwave properties of $Sr_{1-2x}K_xNb_2O_6$ (SNN) is rarely seen in literature, and is hence taken up in the present study.

## Experimental Procedure

Raw materials of high purity $SrCO_3$, $Na_2CO_3$, $Nb_2O_5$ are weighed in accordance with chemical composition of ($Sr_{0.80}Na_{0.40}Nb_2O_6$). The starting raw materials were mixed in a mortar with a pestle for 1 hour, dried and calcined at 1000 $^0$C for 4 hours. The powder was milled again with organic binder polyvinyl alcohol (PVA). It was pressed in to disc type and ring type specimens in suitable die of specific dimensions under pressure. It is then sintered at 1270 $^0$C for 4 hours in

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air. The sintered samples were ground and polished before microwave measurements. X-ray diffraction patterns are taken to study the polycrystalline structure of the ceramic compositions. Analysis of the microstructural features of the composition is done using Scanning electron microscopy (SEM) photographs.

The resonant frequency and Q-factor were measured in the S-band using cavity perturbation method. The measurements were performed by inserting a small pellet sample into a rectangular cavity excited in the TE mode. The properties of the compositions were determined from the resultant change in the frequency and Q-factor. For the measurement of temperature coefficient of resonant frequency Courtney [10] method was used. The temperature coefficient of resonant frequency $f_o$ was measured in the range of 27 to 100 $^0$C. A dielectric resonator of (DR) with the diameter d = 16mm and length l = 6 mm was used for experiment. According to this method, different modes generated were classified and then $\varepsilon_r$ was accurately calculated from $TE_{onl}$ mode. The quality factor Q is determined from the 3 dB bandwidth of $TE_{011}$ mode.

**Results and Discussion.**

Figure 1 shows the X-ray diffraction pattern of $Sr_{1-2x}Na_xNb_2O_6$ ceramics sintered at 1270$^0$C. $Sr_{0.75}K_{0.5}Nb_2O_6$ composition has well defined peaks which are similar to tetragonal tungsten bronze type structure(TTB). The reflections were indexed on the basis of tetragonal symmetry. The calculation of lattice parameters from the d-values gives a = 12.403 A$^0$, c = 3.922 A$^0$. The axial ratio of $\sqrt{10}$ c/a is close to unity, which is characteristic of tetragonal tungsten bronze structure.

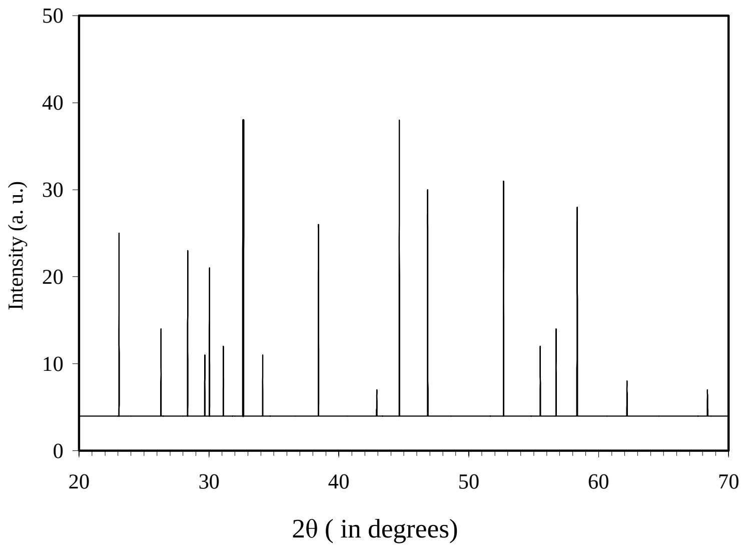


Figure 1. X-ray powder diffraction pattern of $Sr_{0.8}Na_{0.4}Nb_2O_6$ ceramics.

$Sr_{0.80}Na_{0.40}Nb_2O_6$ is a polycrystalline ferroelectric ceramic having microstructure which can be observed by scanning electron microscopy. These microstructures consist of two inter penetrating, phases, the solid and the pore networks. The pores in the ceramic are connected to each other and the pore size distribution is comparably high. The characteristic pore structure and spacing depends on the method of preparation and can range between 2μm – 5 μm. The dimensions of the grains and porous spaces can be calculated from scanning electron micrographs of $Sr_{0.80}Na_{0.40}Nb_2O_6$ sample shown in figure 2. These porous spaces are of much advantage, when we deal with sensing applications. These materials can absorb moisture or water into the porous spaces and are potential materials for microwave sensors and other related applications [8].

The dielectric parameters $\varepsilon_r^{/}$ and $\varepsilon_r^{//}$ of the ceramic materials are obtained from the measured resonance frequencies $f_o$, $f_s$ and the quality factors $Q_o$ and $Q_s$ for the $TE_{10p}$ resonant mode. The complex permittivity (ε′ and ε′′) of the sample can then be calculated from these parameters using microwave cavity perturbation theory [11].

$$\varepsilon_r^{'} - 1 = \frac{f_o - f_s}{2 f_s}\left(\frac{V_c}{V_s}\right) \tag{1}$$

$$\varepsilon_r'' = \frac{V_c}{4V_s}\left(\frac{Q_o - Q_s}{Q_o Q_s}\right) \qquad (2)$$

where $f_o$ is the resonant frequency of the air filled cavity (empty) $f_s$ is the resonant frequency with the sample loaded in the cavity , $V_c$ and $V_s$ are the volume of cavity and sample respectively. $Q_o$ and $Q_s$ are the quality factors of the empty cavity and that of the sample loaded cavity. Figure 3 shows the real part of permittivity of SNN sample calculated using cavity perturbation theory, in the S-band frequency region. The real part of Permittivity is found to vary with frequency. Figure 4 shows the variation of imaginary part of permittivity with frequency.

For the measurement of temperature coefficient of resonant frequency, the cavity was placed in a hot chamber in which the temperature was stabilized. The resonant frequency and line width of the $TE_{011}$ resonance measured when the mode become stationary at each value of temperature. $T_f$ was calculated using the following equation [2].

$$T_f = \frac{1}{f_o}\frac{\Delta f_o}{\Delta T}\Big|_{T=T_o} \qquad (1)$$

The dependence of the relative dielectric constant $\varepsilon_r$ with temperature is specified by the coefficient $T_{\varepsilon}$,which can be calculated using the equation (2).

$$T_\varepsilon = \frac{1}{\varepsilon_r}\frac{\Delta\varepsilon_r}{\Delta T}\Big|_{T=T_o} \qquad (2)$$

Where $f_o$ is the resonant frequency at a given temperature and $\Delta f = f_0(T) - f_0$ (27 $^0$C). The temperature coefficient of resonant frequency, $\tau_f$ is the parameter which indicates the thermal stability of the resonator. The $T_f$ was found to be very small, equal to 1.46137 ppm /$^0$C at 9.37GHz. Figure 5 shows the temperature variation of dielectric constant measured using Courtney method. The variation in dielectric constant is found to be negligibly small when temperature increases from 27$^0$ C to 100$^0$ C. The dielectric constant comes out to be a linear function of temperature as shown is figure 5. For the reference temperature 60$^0$ C, the temperature coefficient of the dielectric constant is computed in accordance with equation (2)

$T_\varepsilon$ = (10.447-10.449) ⁄ [30.448 × (100 -27)] = -2.622 ppm/$^0$C

Due to the linear behaviour, the same coefficient is valid over the entire range of temperature.

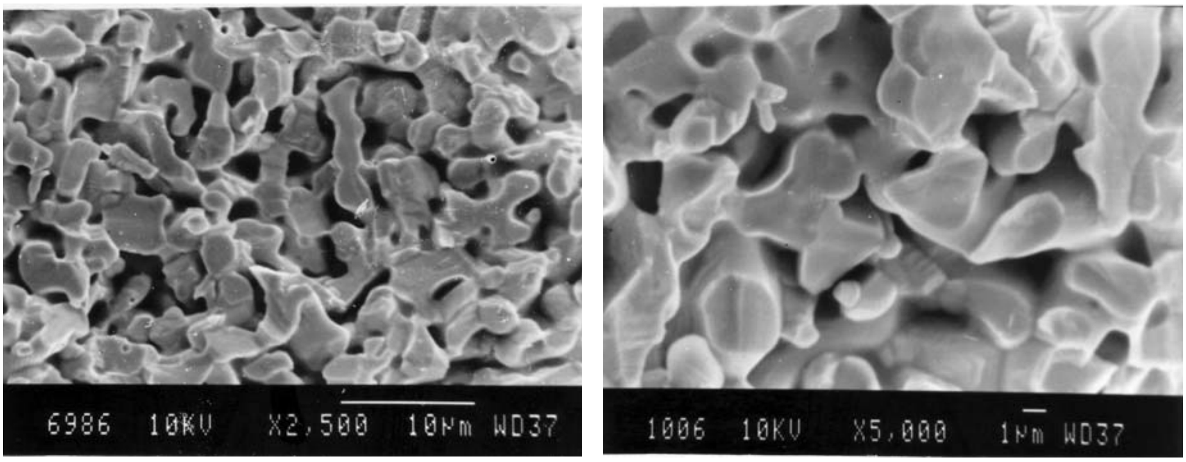


Figure 2 Scanning electron microscope photographs of SNN ceramics.

The variation of the resonant frequency with temperature is described in terms of physical parameters, one part is caused by the physical expansion of material parts, and the other by the change in the relative dielectric constant. The dielectric constant and temperature coefficient of resonant frequency depend mainly on the composition of the material. But loss factor varies from sample to sample of the same composition, indicating that the loss factor is sensitive to slight differences in crystallographic structure and microstructure of the material. One of the loss mechanisms in dielectric materials is the

resonance of electric dipoles in the molecule of the material. At a particular frequency, the dipole motion becomes resonant and will absorb power from the exciting electromagnetic field and the permittivity of the material is reduced.

Figure 6 shows the conductivity (S/m) variation of strontium barium niobate compositions with frequency. Conductivity is found to be varying with frequency in the S-band spectral region. The reflection characteristics of a DR made of SNN and fed from a coaxial feed are obtained. The dielectric resonator is having a bandwidth of 675 MHz at the resonant frequency 1855 MHz. The material exhibits good characteristics suitable for use as dielectric resonators and DR antennas.

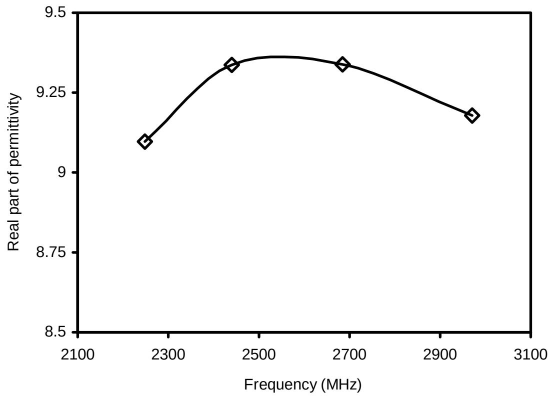


Figure 3. Frequency versus real part of complex Permittivity of SNN ceramics.

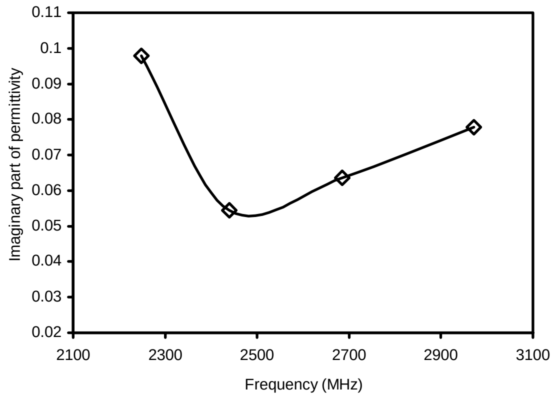


Figure 4. Frequency versus imaginary part of complex Permittivity of SN ceramics.

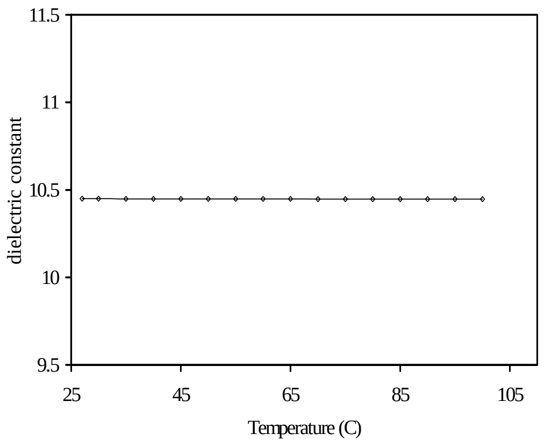


Figure 5. Variation of dielectric constant with temperature for SNN ceramics.

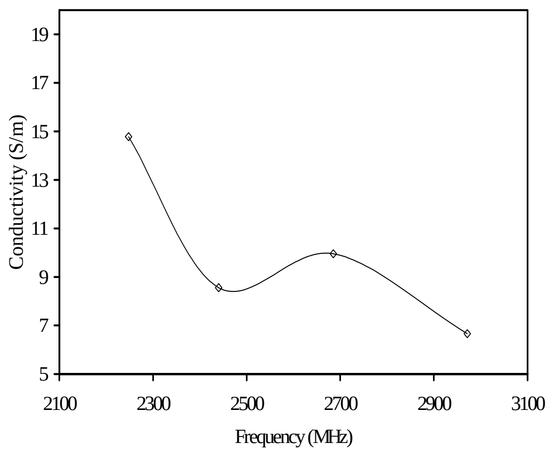


Figure 6. Frequency versus conductivity (S/m) of SNN ceramics.

## Conclusion

Strontium sodium niobate, $Sr_{0.80}Na_{0.40}Nb_2O_6$, prepared by solid state reaction method showed tetragonal tungsten bronze type structure. The microstructural study shows the porous polycrystalline features of this composition. The microwave complex permittivity and conductivity are studied as a function of frequency. The temperature coefficient of dielectric

constant and resonant frequency is found to be very small. The material exhibits good characteristics suitable for use as dielectric resonators and DR antennas.

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